
Removal of hexavalent chromium using coffee husk

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Abstract: The potential to remove chromium(VI) from aqueous solutions through biosorption using coffee husk was investigated. The effects of pH, contact time, initial concentration and adsorbent dosage on the adsorption of Cr(VI) were studied. The data obeyed Langmuir and Freundlich adsorption isotherms. The Langmuir adsorption capacity was found to be 44.95 mg/g. The Freundlich constants K_f and n were 1.027 [mg/g (l/mg)ⁿ] and 1.493, respectively. Desorption studies indicated the removal of 60% of the hexavalent chromium. Infrared spectral studies revealed the presence of functional groups, such as hydroxyl and carboxyl groups, on the surface of the biomass, which facilitates biosorption of Cr(VI).

Keywords: chromium(VI); coffee husk; low-cost biosorbent; adsorption; infrared analysis.

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1 Introduction

Chromium is an essential nutrient for plant and animal metabolism. However, the increasing accumulation of chromium in the environment from industrial outputs has caused great concern. Chromium-contaminated wastewaters can originate from dyes and pigment manufacturing, wood preserving, electroplating and leather tanning. Chromium exists in +3 and +6 oxidation states, as all other oxidation states are not stable in aqueous solutions. Both valences of chromium are potentially harmful (Dakiky et al., 2002). Hexavalent chromium which is primarily present in the form of chromate (CrO_4^{2-}) and dichromate ($\text{Cr}_2\text{O}_7^{2-}$) poses significantly higher levels of toxicity than the other valency states (Sharma and Forster, 1995).

Conventional methods for removing Cr(VI) ions from industrial wastewater include reduction (Kim et al., 2002), reduction followed by chemical precipitation (Ozer et al., 1997), adsorption on activated carbon (Lotfi and Adhoum, 2002), solvent extraction (Mauri et al., 2001), freeze separation, reverse osmosis (Padilla and Tavani, 1999), ion exchange (Rengaraj et al., 2003) and electrolytic methods (Namasivayam and Yamuna, 1995). These methods have found limited application because they often involve high capital and operational costs. Adsorption is an effective and versatile method for removing chromium. Natural materials that are available in large quantities or certain waste products from industrial or agricultural operations may have potential as

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inexpensive sorbents. Most of the low-cost sorbents have the limitation of low sorptive capacity and thereby for the same degree of treatment, it generates more solid waste (pollutant-laden sorbent after treatment), which poses disposal problems. Therefore, there is a need to explore low-cost sorbents having high contaminant sorption.

Fly ash from thermal power plant (Panday et al., 1984), waste slurry from a fertiliser plant (Srivastava et al., 1989) and Fe(III)/Cr(III) hydroxide obtained from the petrochemical industry (Namasivayam and Ranganathan, 1993), blast furnace flue dust (Patnaik and Das, 1995) and photo film waste sludge (Selvaraj et al., 1997) have been examined for the removal of hexavalent chromium. The adsorption of Cr(VI) on bituminous coal (Rawat and Singh, 1992), sphagnum peat moss (Sharma and Forster, 1993), coconut husks and palm pressed fibres (Tan et al., 1993), sawdust, sugarcane bagasse, sugarbeet pulp and maize cob (Sharma and Forster, 1994) has been reported. A thorough literature survey indicated that coffee husk has not been used as an adsorbent thus far. The objective of the present study was to explore the feasibility of using coffee husk to remove hexavalent chromium from aqueous solutions. The effects of pH, contact time, biomass concentration and adsorption equilibrium were investigated.

2 Materials and methods

The coffee husk was collected from coffee processing unit. The husk was washed and boiled to remove colour and impurities. The biomass was then air dried at 105 °C for 24 hours and the adsorbent thus processed was used in its original piece size. A stock solution containing 1000 mg/l of Cr(VI) was prepared using potassium dichromate in distilled water. All the chemicals used were of analytical grade. Batch mode experiments were conducted by agitating 1 g of the adsorbent with 100 ml of Cr(VI) solution of desired concentration at pH 2. The adsorbent was separated using Whatman No.1 filter paper and the supernatant was analysed spectrophotometrically using 1,5-diphenylcarbazide (APHA, 1985). To study the effect of pH, it was varied between 2 and 7 at different initial metal concentrations. The pH was adjusted using 0.1 N NaOH and 0.1 N HCl. The effect of adsorbent dosage was studied by varying the adsorbent from 0.5 g to 6 g at various initial metal concentrations at pH 2. The desorption studies were carried out by filtering the Cr(VI) loaded adsorbent from solution, washing gently with distilled water to remove unadsorbed Cr(VI) and agitating the metal loaded adsorbent with distilled water and varying concentrations of NaOH and HCl. Infrared spectroscopic studies of coffee husk before and after treatment with Cr(VI) was carried out to determine the functional groups responsible for adsorption using a Fourier Transform Infrared Spectrometer (FTIR).

3 Results and discussion

3.1 Characteristics of the adsorbent

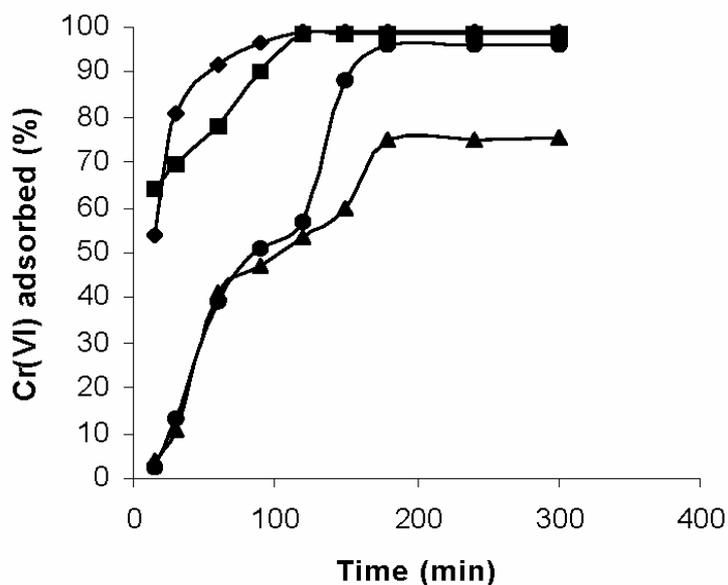
The carbon, hydrogen and nitrogen percentages of the coffee husk were 45.33, 6.21 and 0.63, respectively. A relatively larger percentage of hydrogen in comparison to nitrogen compounds indicates that carbon-hydrogen groups might be available for adsorption of

metals. The relatively low percentage of nitrogen shows that very less percentage of protein might be present in the husks. This is advantageous over protein-rich adsorbents since proteinous materials are likely to putrefy under moist conditions.

3.2 Effect of agitation time and initial Cr(VI) concentration on Cr(VI) adsorption

The effect of agitation time for various initial concentrations was studied (Figure 1). The percentage adsorption of Cr(VI) increased with an increase in agitation time and the equilibrium time varied for different Cr(VI) concentrations. The time required to attain equilibrium for 10 and 20 mg/l Cr(VI) concentration was 120 minutes and 50 and 100 mg/l Cr(VI) attained equilibrium at 180 minutes. The maximum percentage adsorption was 99%, 98%, 96% and 75% respectively for initial Cr(VI) concentrations of 10, 20, 50 and 100 mg/l.

Figure 1 Effect of agitation time and initial Cr(VI) concentration (◆, 10 mg/l; ■, 20 ppm; ●, 50 mg/l; ▲, 100 mg/l) on Cr(VI) removal (adsorbent dose: 1 g/100 ml; pH 2.0)

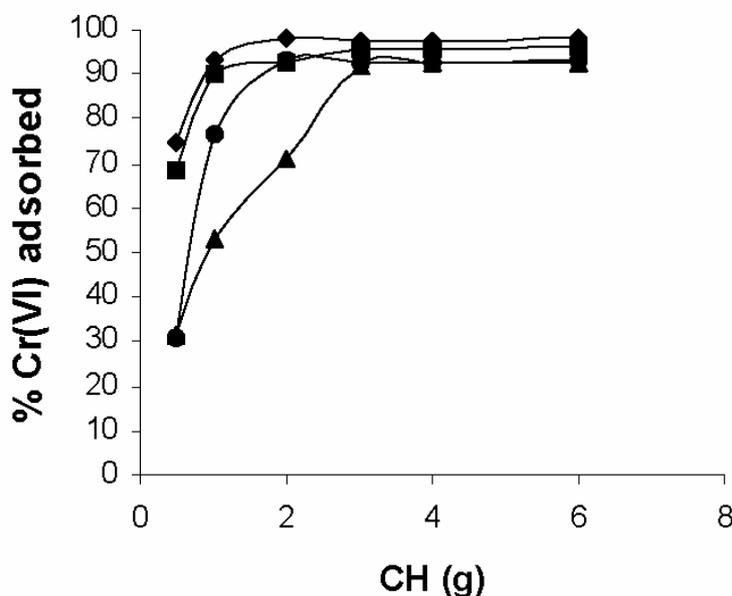


3.3 Effect of adsorbent dose on adsorption

The percentage adsorption of Cr(VI) was studied by increasing adsorbent dose from 50 mg to 6 g for 100 ml of Cr(VI) concentration of 10, 20, 50 and 100 mg/l (Figure 2). The results indicated that the percentage of Cr(VI) adsorbed increased with an increase in adsorbent dosage for all Cr(VI) concentrations. The increase in percentage adsorption with increase in adsorbent dosage is due to the increase in the number of adsorption sites (Sharma and Forster, 1993; Selvaraj et al., 1997).

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Figure 2 Effect of adsorbent dose on Cr(VI) adsorption at various Cr(VI) concentrations (◆, 10 mg/l; ■, 20 ppm; ●, 50 mg/l; ▲, 100 mg/l; agitation time 180 minutes; initial pH 2.0)



3.4 Effect of pH

The pH of the solution is an important factor that controls the uptake of Cr(VI). The experimental results revealed that the percentage adsorption increased as the pH was lowered and reached 99%, 98% and 96% respectively for 10, 20 and 50 mg/l Cr(VI) concentrations. When the pH was increased above 2, the per cent removal decreased (Figure 3). Similar results were reported by Donmez and Aksu (2002), Dakiky et al. (2002), Selvaraj et al. (2003), Yu et al. (2003), Uzun et al. (2002), Hu et al. (2003) and Gupta et al. (2001). The high adsorption of Cr(VI) can be explained by the species of chromium and the adsorbent surface. At acidic pH, the predominant species of Cr(VI) are $\text{Cr}_2\text{O}_7^{2-}$, HCrO_4^- and CrO_4^{2-} . Under acidic conditions, the surface of the adsorbent becomes protonated and attracts anionic species of Cr(VI). As the pH is increased above the zeta potential of the adsorbent, there is a reduction in the electrostatic attraction between the Cr(VI) species and the adsorbent surface, with a consequent decrease in percentage adsorption.

3.5 Adsorption isotherms

The Langmuir isotherm was applied to the present study to estimate the adsorption capacity of coffee husk. It is valid for monolayer adsorption onto a surface containing a finite number of identical sites (Langmuir, 1918). The linear plots of C_{eq}/q vs C_{eq} for Cr(VI) show that adsorption follows the Langmuir adsorption model (Figure 4). q_{max} and b were determined from the slope and the intercept of the plot to be 44.95 mg/g and 0.01 l/mg, respectively.

Figure 3 Effect of pH on Cr(VI) adsorption at different initial Cr(VI) concentrations (◆, 10 mg/l; ■, 20 ppm; ●, 50 mg/l; ▲, 100 mg/l)

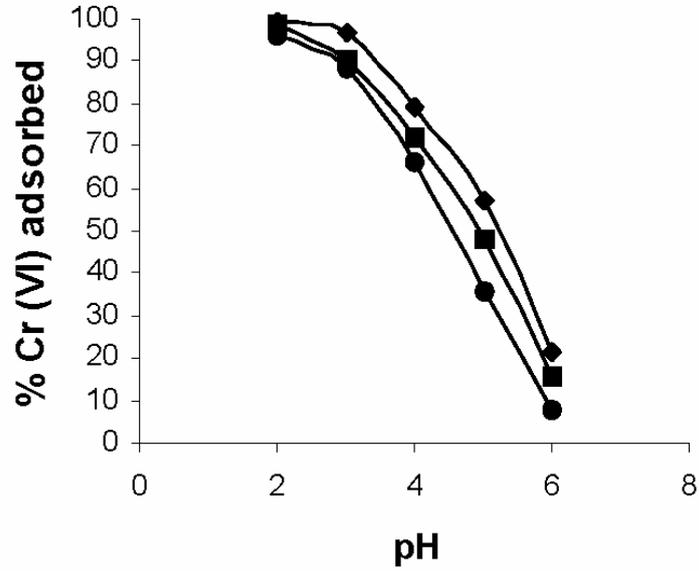
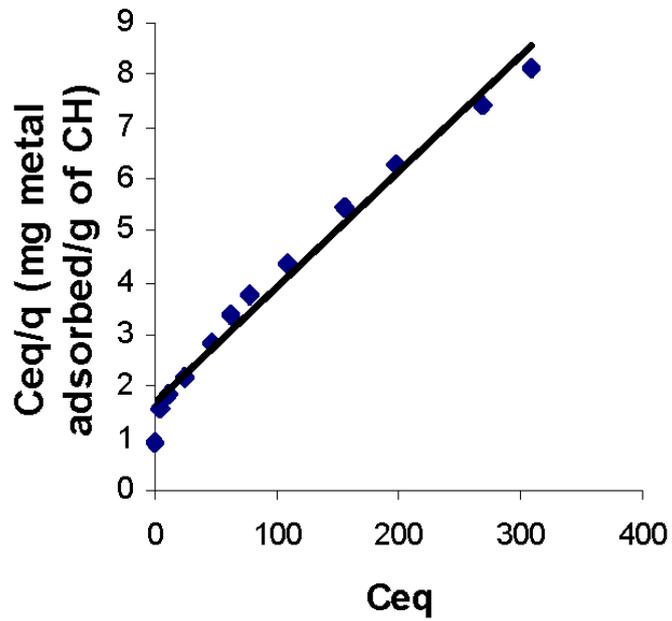


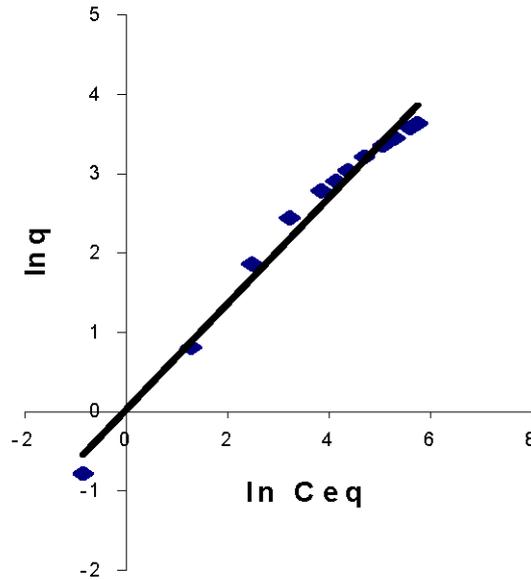
Figure 4 Langmuir plot for Cr(VI) adsorption (agitation time 180 minutes; initial pH 2.0) (see online version for colours)



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The Freundlich adsorption isotherm (Freundlich, 1907) was also applied for the adsorption of Cr(VI) onto coffee husk. Linear plots of $\ln C_{eq}$ vs $\ln q$ show that the adsorption of metal ions onto the coffee husk follows the Freundlich isotherm model (Figure 5). Freundlich constants K_f and n were found to be 1.02 mg/g and 1.49, respectively. It has been shown by McKay et al. (1982) that n values between 1 and 10 indicate beneficial adsorption.

Figure 5 Freundlich plot for Cr(VI) adsorption (initial pH: 2.0; agitation time 180 minutes) (see online version for colours)



The adsorption data obeyed both Freundlich and Langmuir models exhibiting heterogeneous surface conditions and monolayer adsorption (Lee et al., 1995). The favourable nature of adsorption can be expressed in terms of a dimensionless parameter R_L given by the equation

$$R_L = 1/(+bC_0) \quad (1)$$

where b is the Langmuir constant and C_0 is the initial concentration of Cr(VI). All R_L values obtained using equation (1) for Cr(VI) adsorption are greater than zero and less than unity showing favourable adsorption of Cr(VI) onto coffee husk.

3.6 Comparison with other adsorbents

The comparison of adsorbent capacity of coffee husk with other materials reported in literature is given in Table 1. The sorption capacity of coffee husk is higher than adsorbents from various industrial and low-cost adsorbents.

Table 1 Comparison of adsorption capacity of Cr (VI) with other adsorbents

<i>Biosorbent</i>	<i>Cr(VI) mg/g</i>	<i>Reference</i>
Distillery sludge	5.7	Selvaraj et al., 2003
Exhausted coffee	1.42	Orhan and Büyüğüngör, 1993
Nut shell	1.47	Orhan and Büyüğüngör, 1993
Sawdust	10.1, 16.05, 4.44	Bryant et al., 1992; Dikshit, 1989; Zarraa, 1995
Walnut shell	1.5	Orhan and Büyüğüngör, 1993
Waste tea	1.63	Orhan and Büyüğüngör, 1993
Irish sphagnum moss peat	119.0, 43.9	Sharma and Forster 1993, 1995
Iron (III) hydroxide	0.47	Namasivayam and Rangnathan, 1993
Blast furnace slag	7.5	Srivastava et al., 1997
Activated red mud	1.6	Pradhan et al., 1999
Rice husk carbon	45.6	Srinivasan et al., 1988
Waste tyre	58.48	Hamadi et al., 2001
Olive cake	33.44	Dakiky et al., 2002
Pine needles	21.50	Dakiky et al., 2002
Wool	41.15	Dakiky et al., 2002
Coffee husk	44.95	This study

3.7 Infrared spectral analysis

The FTIR spectral analysis shows that several functional groups are available on the surface of coffee husk for binding Cr(VI) ions (Table 2).

Table 2 Wavelengths corresponding to their respective functional groups

<i>Wavelength (cm⁻¹)</i>	<i>Functional group</i>
3431	–OH
2925	–CH
1726	C=O
1652	C=C
1430	–COO

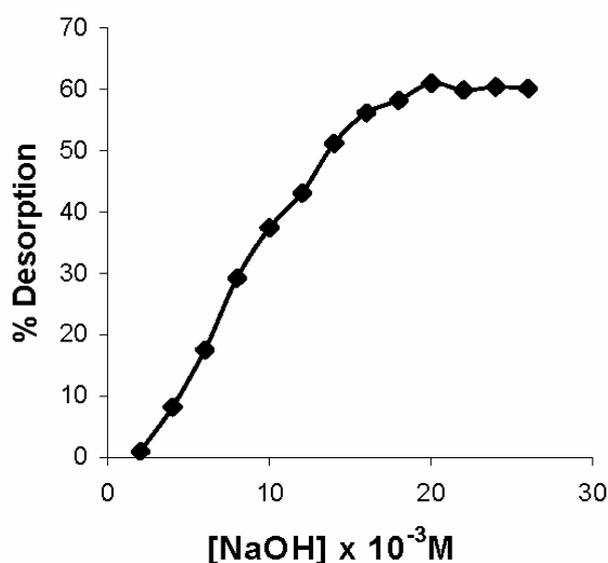
3.8 Desorption studies

Adsorption process can either be physisorption (physical bonding) or chemisorption (chemical bonding) or combination of both. If the adsorption is by physical bonding then the loosely bound metal ion can be easily desorbed with distilled water in most of the cases. However, if the mode of sorption is by chemical bonding or ion exchange or combination of both, then desorption can be effected by stronger desorbents like acid or alkali solutions. Distilled water (pH 6.9) was used to desorb Cr(VI) from Cr(VI) loaded coffee husk. The results revealed that the Cr(VI) was not removed from metal-laden biomass. Hence the experiments were conducted with acid and alkali solutions. The Cr (VI) ions were desorbed with alkali solutions (NaOH) and not with acid

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solutions. Figure 6 shows that about 60% of Cr(VI) was desorbed using 0.02 M NaOH. From the desorption experimental results it can be concluded that the mode of biosorption of Cr(VI) on coffee husk was chemisorption. The results of alkali desorption of metal suggests either chemisorption or ion exchange as the possible mechanism of binding between Cr(VI) and coffee husk. At higher pH, hydroxyl ions may release chromium ions from the adsorbent following an ion exchange mechanism.

Figure 6 Effect of NaOH concentration on desorption of Cr(VI)



4 Conclusion

The experimental results show that coffee husk is an excellent alternative for the removal of Cr(VI) from aqueous solutions. The biosorption of Cr(VI) was dependent on pH, contact time, adsorbent dosage and initial metal concentration. Both Langmuir and Freundlich isotherms were followed by the adsorption of Cr(VI). The infra red spectral analysis showed that the functional groups like carboxyl and hydroxyl ions played an important role in the biosorption of Cr(VI). As the coffee husk is easily available from coffee processing units, its utility as biosorbent will be economical and be viewed as part of waste management strategy. Containing very less amount of protein ($N = 0.63$), coffee husk is advantageous over the protein-rich algal and fungal biomass projected as metal biosorbents, since proteinous materials are likely to putrefy under moist conditions. Moreover, the desorption experiments show that the metals can be desorbed after adsorption and Cr(VI) can be recovered. The present adsorbent can be used at an industrial scale to remove chromium ions from the effluents before discharging into the environment. Coffee husk can replace the expensive activated carbon in the adsorption process. Most of the electroplating effluents contain chromium as one of the major contaminant, which can be removed in a cost-effective and efficient manner by coffee husk.

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